Orbital weight redistribution triggered by spin order in the pnictides

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The one-particle spectral function and its orbital composition are investigated in a three-orbital model for the undoped parent compounds of the iron-based superconductors. In the realistic parameter regime, where results fit experimental data best, it is observed that the magnetization in the xz and yz orbitals are markedly different and the Fermi surface presents mostly xz character, as recently observed in photoemission experiments [T. Shimojima *et al.*, Phys. Rev. Lett. **104**, 057002 (2010)]. Since the ferro-orbital order in this regime is at most a few percent, these results are mainly driven by the magnetic order. An analogous analysis for a five-orbital model leads to similar conclusions.

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Introduction. In contrast to the cuprate superconductors, more than one band cross the chemical potential in the pnictide superconductors.[1]According to density functional theory (DFT) calculations, most of the spectral weight at the Fermi surface (FS) arises from two of the iron d-orbitals, namely the xz and yz orbitals (inset of Fig. 1), which are degenerate in the high-temperature tetragonal phase. Lowering the temperature, the undoped parent compounds undergo a structural as well as a magnetic phase transition to an orthorhombic phase with antiferromagnetic (AF) order with wavevector $(\pi, 0)$[2](inset of Fig. 1). This spin order breaks the rotational symmetry of the original tetragonal lattice. In this regime, scanning tunneling microscopy[3] and resistivity measurements[4] have indicated that the electronic system presents symmetry-breaking properties that far exceed the relatively modest difference of the lattice constants before and after the transition.

To rationalize these results, orbital ordering has been suggested to occur together with the magnetic ordering, lifting the degeneracy between the xz and yz orbitals and inducing the lattice distortion.[5–7] Such a ferro-orbital (FO) ordered state has anisotropic magnetic couplings, which stabilize the AF order without necessarily frustrating any magnetic interactions,[5,7,8] thus explaining the spin-wave dispersions which do not indicate frustrated AF couplings.[9] However, while clear AF signatures have been observed in Angle Resolved Photoemission (ARPES),[10,11] the situation is less clear for features due to orbital ordering. DFT results have been interpreted in terms of FO order,[7] but a detailed account of the impact of orbital order on the spectral properties is still lacking. Finally, ARPES results can be fitted by DFT if the magnetic moment in the calculation is artificially suppressed towards experimentally observed values, and these calculations then yield a far smaller rearrangement of the two hole pockets,[11] indicating that FO order and the ordered magnetic moment may be linked.

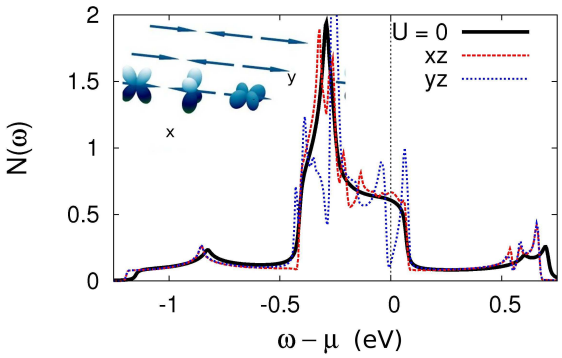

FIG. 1: (Color online) Density of states of the xz and yz orbitals in the $(\pi, 0)$-AF phase of the three-orbital model at $U = 0.7$ and $J = U/4$. For these values of U and J, the orbital densities are $n_{xz} = n_{xz,\uparrow} + n_{xz,\downarrow} = 1.590 \approx n_{yz} = 1.586$, and the magnetizations $m_{xz} = n_{xz,\uparrow} - n_{xz,\downarrow} = 0.04 \ll m_{yz} = 0.15$. For $U = 0$, $N(\omega)$ is identical for both orbitals (solid line). A Gaussian broadening with $\sigma = 0.005$ was used. The inset illustrates the $(\pi, 0)$-AF order considered here and the xz, yz, and xy orbitals (left to right).

In this paper, the spectral function of a three-orbital model[8] is investigated with an emphasis on features related to orbital polarization effects.This model allows for the stabilization of a regime with small ordered magnetic moments[12,13] by selecting intermediate values for the Hubbard repulsion. One-particle spectral functions calculated in this regime have already been shown to be qualitatively similar to experimental ones,[8] including the presence of small hole- or electron-like extra pockets near the electron and hole pockets of the uncorrelated bands.[11,14] We focus here on the orbital composition of the Fermi surface, which was not analyzed in those previous investigations. It will be shown that the FS has predominantly xz character, similar to recent experimental results obtained with Laser-ARPES.[15] In this regime

with the polarized FS, the orbital magnetizations show a substantial difference between the yz and xz orbitals, but there is hardly any static orbital order. Our analysis of the three-orbital model is complemented by a discussion of a five-orbital model,[16] where similar results are found. This model admits a regime with moderate FO order of $\approx 30\%$; the spectral density and FS, however, more closely resemble ARPES results if the FO is at most a few percent and the ordered magnetic moment is small or intermediate.[17]

Model. The Hamiltonian studied here consists of the kinetic energy (tight binding) previously used for three[8] or five[16] d orbitals, as well as the standard onsite Coulomb interaction terms comprised of the intraorbital repulsion U, interorbital repulsion U', and the z-component of the Hund's rule interaction regulated by a coupling J, with $U = U' + 2J$. The reader is referred to Refs. 8,16 for more details. The overall electronic density per site n is 4 (6) for the three (five) orbital model. The spin-flip and pair-hopping terms, which are by symmetry also part of the onsite interaction, drop out in our previous and current mean-field studies.[18] The interacting Hamiltonian is then treated with a mean-field approximation,[12] where we can compare a variety of phases with different magnetic and orbital orders.[8] For a two-orbital model, our method was compared to the Variational Cluster Approximation,[19] and found to give similar results.[12,13] As previously reported, small to intermediate Coulomb repulsions $\lesssim$ 1eV stabilize an AF metal in agreement with experiments.[8,12] Approximations beyond mean-field will likely increase the actual values of U and J in the realistic regime.

Results for three orbitals. Figure 1 shows the xz and yz contributions to the density of states, both for the AF metal found at $U = 0.7$ and $J = U/4$, and for the uncorrelated system. In agreement with the interpretation given to Laser-ARPES results in Ref. 15, we find that most of the weight at the FS arises from the xz orbital in the AF state, while both orbitals contribute equally in the tetragonal nonmagnetic state. The total densities in the xz and yz orbitals are, however, almost the same $n_{xz} = 1.590 \approx n_{yz} = 1.586$, i.e., there is (almost) no FO order. Only the states near the chemical potential μ are xz-polarized, and a strong yz peak at energies ≈ -50 meV approximately compensates for the missing yz-weight around μ. This peak comes from the opening of a gap that stabilizes the AF order and that affects mainly the yz portions of the bands. The system remains metallic, because the xz orbital does not have a gap around μ. The stronger impact of the magnetic $(\pi, 0)$-order on the yz orbital also leads to a larger magnetization for this orbital, with $m_{yz} = 0.15 \gg m_{xz} = 0.04 \gg m_{xy} = 0.014$. Such a larger value of m_{yz} has been explained by a larger yz hopping along the x-direction,[7] but in the present three-orbital model the xz orbital has the largest hopping amplitude along x.[20] While we have observed before a dominant m_{xz} in the large-U limit,[8] we attribute the relatively large value of $m_{yz} > m_{xz}$ observed at smaller

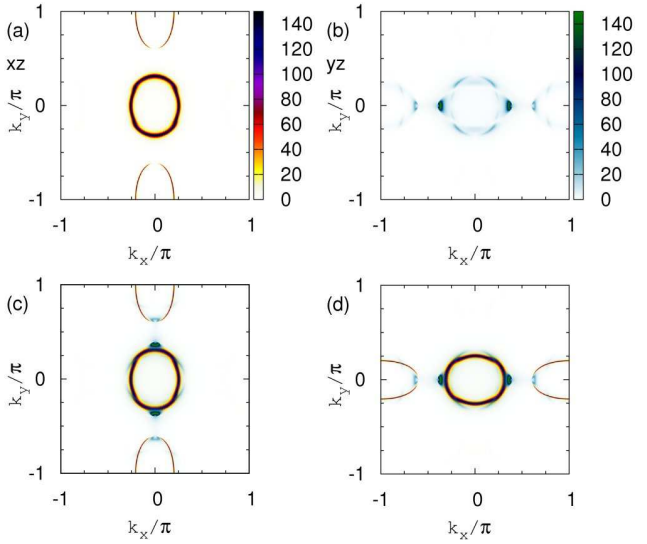

FIG. 2: (Color online) Contributions of the (a) xz and (b) yz orbitals to the Fermi surface for the three-orbital model at the same parameters as in Fig. 1. (c) and (d) show the superpositions of xz and yz as they would be expected to appear in the s and p polarizations, see text. The FS also has a small weight in the xy orbital at the "tip" of the electron pocket (not shown).

U to the orbital character of the electron pockets. The pocket found around $(\pi, 0)$ in the uncorrelated model, with mostly yz character, gets folded into the central hole pockets and forms the gap mentioned above, while the xz pocket at $(0, \pi)$ is far less affected.[21] Since the yz orbital develops a pseudogap at μ and the ungapped xz orbital consequently determines the states at the Fermi level, the slightly higher resistivity in the ferromagnetic (FM) y-direction[4] might be due to the fact that the xz orbital has larger hopping amplitudes in the AF x-direction.

Figures 2(a) and (b) show the xz and the yz contributions to the Fermi surface for the same parameters as in Fig. 1. As mentioned above, and as also reported previously,[17] most of the FS is given by xz states, but we also find small features coming from the yz orbital. These small yz electron-like pockets, see also Fig. 6(a) in Ref. 8, are similar to V-shaped features reported in Laser-ARPES[15] and their yz character does not contradict the experimental findings: the laser spot is expected to catch signals both from $(\pi, 0)$- and $(0, \pi)$-ordered domains, and the two polarizations pick up either the xz or the yz orbital. Since the yz orbital takes the same role for $(0, \pi)$ that xz has for $(\pi, 0)$, the polarization sensitive to xz symmetry is expected to find states with xz character from $(\pi, 0)$ domains together with features having what corresponds effectively to 'yz' from the rotated $(0, \pi)$ domains. Similarly, changing the polarization leads to yz for $(\pi, 0)$ plus 'xz' for $(0, \pi)$.[15] This situation can be modeled by adding to the xz-weight at the FS the yz contributions rotated by 90 degrees, because they stem from domains with rotated AF order. Figures 2(c) and 2(d)

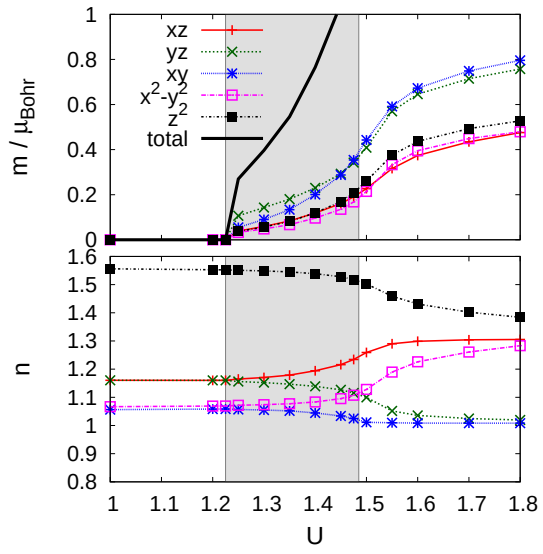

FIG. 3: (Color online) Orbital dependent (a) magnetizations (in μ_B) and (b) electronic densities of the five-orbital model varying the strength of the Coulomb repulsion U, at $J = U/4$. In (a), the total magnetization is also given. The gray area denotes the regime with small ordered magnetic moment and almost no FO order. Its lower boundary is given by the onset of a finite ordered moment. Since orbital order develops more gradually, its upper boundary was defined as the inflection point of the orbital densities, which coincides well with an inflection point in the magnetization.

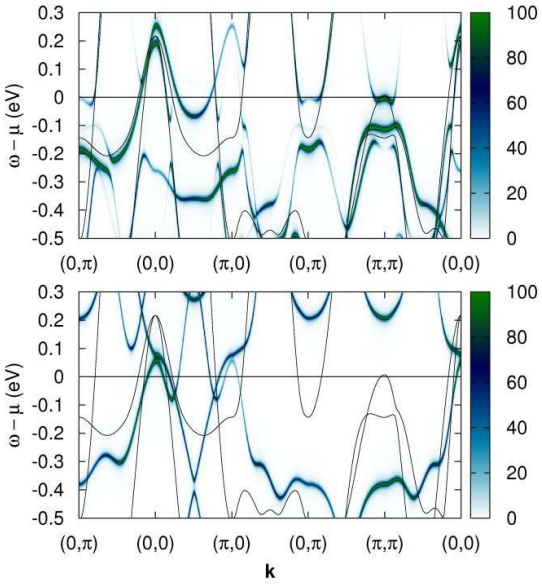

FIG. 4: (Color online) One-particle spectral function $A(\mathbf{k},\omega)$ for (a) the AF metal without orbital order and (b) the AF metal with moderate FO order at $U = 1.6$. In (a), for $U = 1.35$ and $J = U/4$, the total staggered magnetization is $m_{\rm tot} = 0.55\mu_B$, and the orbital contributions of the xz and yz orbitals are $m_{xz} = 0.08\mu_B$ and $m_{yz} = 0.18\mu_B$. The difference in the densities is $n_{xz} - n_{yz} = 0.032$. In (b), for $U = 1.6$ and $J = 0.4$, $m_{\rm tot} = 2.52\mu_B$, $m_{xz} = 0.37\mu_B$, and $m_{yz} = 0.65\mu_B$; the difference in the densities is $n_{xz} - n_{yz} = 0.26$. The thin black lines give the uncorrelated bands at $U = 0$.

show the expected result for the two polarizations, and one clearly observes the rotation of all features by 90 degrees, as seen in experiments. The rotation would only break down for a hybridized FS that contains substantial contributions from both the xz and yz orbitals. Both hole pockets present such a mixture in the uncorrelated bands,[8] but Figs. 2(a) and (b) clearly show that the AF order removes the hybridization and each feature of the FS in the AF phase has (almost) only xz *or* yz character.

Results for five orbitals. A similar analysis was carried out for a five-orbital model[16] and Fig. 3 shows the mean-field results for the magnetizations and densities in the five d-orbitals, varying the strength of the on-site Coulomb repulsion U.[22] A realistic constant ratio $J = U/4$ was chosen, and it was checked that a slightly larger or smaller J does not qualitatively alter our conclusions. For small U, the system remains an uncorrelated metal without any magnetic ordering and practically unchanged orbital densities. As was reported for the other multi-orbital models,[8,12] AF order starts to develop at a critical value of U, see Fig. 3(a). The staggered magnetization per site, which corresponds to the ordered magnetic moment, grows continuously in this regime and remains smaller than $1.5\mu_B$.

Similarly as for the three-orbital model, the five-orbital model is away from half-filling, and orbital ordering effects could therefore occur more easily than in the half-filled four- and two-orbital models.[23,24] However, again similarly as for the three-orbital model, the first critical U turns out not to affect the orbital densities as strongly as the magnetization, see Fig. 3(b). The densities only slowly begin to vary after a robust magnetization has set in and for a finite window in U the difference remains in the low percent range, far smaller than the magnetization. Moreover, the difference in orbital densities is also smaller than the difference in orbital magnetizations with $m_{yz} > m_{xz}$, due to the yz band being more strongly gapped around the chemical potential (see the discussion for the three-orbital model above). Only for larger values of U, where the ordered magnetic moment is already quite large, a moderate FO order sets in with $\approx 30\%$ more electrons in the xz orbital. Even in this phase, we find that all orbitals are affected to a similar degree, even though the xz and yz orbitals, which are degenerate in the uncorrelated case and make up most of the weight at the FS, might *a priori* be expected to be particularly susceptible to symmetry-lowering orbital order.

Figure 4(a) gives the spectral function $A(\mathbf{k},\omega)$ for $U = 1.35$ and $J = U/4$, where the AF ordered magnetic moment is relatively small $m_{\rm tot} = 0.55\mu_B$ and where the densities in the xz and yz orbitals differ only by a few percent i.e., in the gray-shaded area of Fig. 3. The spectral function is rather similar to the uncorrelated one, but some gaps have opened, at the chemical potential (mostly for the yz orbital) and also away from it. The

resulting "shadow" bands of magnetic origin[25] form additional hole-pocket–like features next to the original electron pockets, in agreement with ARPES.[14] Similar to the three-orbital model discussed above, xz-yz-hybridization of the hole pockets has given way to a large xz-polarized central pocket and small satellites with yz character. If a slightly larger $U = 1.6$ is chosen, so that the system develops some FO order, see Fig. 3(b), the spectral function changes considerably, as it can be seen in Fig. 4(b): all remnants of the original electron pockets have completely disappeared, there are no longer bands just below the chemical potential around $(\pi, 0)/(0, \pi)$ as seen in ARPES,[11] and the features around $(0, 0)$ and $(\pi, 0)$ are far more symmetric to each other than in ARPES.[11,14] The strong reconstruction into features that resemble neither the uncorrelated bands nor ARPES experiments arises because the interaction is now strong enough to involve the states at (π, π), located just below the chemical potential at $U = 0$, in the magnetic order.

Conclusions. In summary, both the three- and five-orbital models have instabilities towards orbitally ordered states, and the instability can be driven by $(\pi, 0)$-AF order. However, significant orbital order requires a relatively strong onsite Hubbard repulsion U and goes together with a large ordered magnetic moment and a significant reconstruction of the one-particle bands and FS, see Ref. 8 and Figs. 3 and 4(b). At small to intermediate U, a realistic AF metal with small ordered magnetic moments is found where the densities in the xz and yz orbitals differ by at most a few percent.

The orbital *magnetization*, on the other hand, is far stronger for the yz orbital [for AF order with ordering vector $(\pi, 0)$] than it is for xz, which suggests that the orbital degree of freedom strongly couples to the magnetic order. Such a more dynamic picture of the orbital degree of freedom in pnictides is also corroborated by the one-particle density of states, where the states near the Fermi surface have more xz character than yz, leading to a FS with substantial orbital polarization, even in a regime where FO order is at most a few percent. Another effect of the magnetic order is the breakdown of the hybridization between the xz and yz orbitals: while the uncorrelated FS shows features with mixed xz-yz character, all features in the correlated FS are either purely xz or, for some smaller pockets, purely yz, in agreement with Laser-ARPES results.[15]

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[20] Not only is the intraorbital hopping in the x-direction smaller for yz than for xz, but the substantial *inter*orbital hopping connecting the yz (xz) orbitals to xy is 0 along x (y) for symmetry reasons.[13]

[21] Since our mean-field approach breaks the orbital symmetry by neglecting the 'pair-hopping' and spin-flip terms of the onsite interaction, we have to check that the yz electron pocket's predominant interaction with the yz parts of the hole pockets is not an artifact of the approximation. We perform the mean-field calculation in a rotated basis with orbitals $|a\rangle = \cos\alpha|xz\rangle + \sin\alpha|yz\rangle$ and $|b\rangle = -\sin\alpha|xz\rangle + \cos\alpha|yz\rangle$[8] and find that (i) the energy is minimal for the original $\alpha = 0$ and that (ii) $m_{yz} > m_{xz}$ persists for $\alpha \neq 0$.

[22] For the intermediate-U regime with small ordered moment, the $(\pi, 0)$ phase has actually slightly higher energy than a (π, π)-AF phase with large ordered moment, but since the five-band model is widely used by several authors, we are nevertheless going to discuss its properties under the assumption of $(\pi, 0)$ order.

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